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A simple hydride model for cerium ejecta particles

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Funding: ASC/PEM/Mix and Burn
ASC/IC/LAP
LDRD

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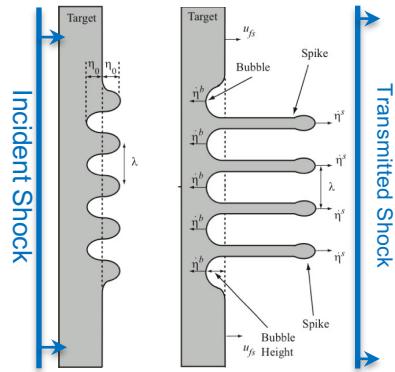
May 20, 2021
XCP Seminar



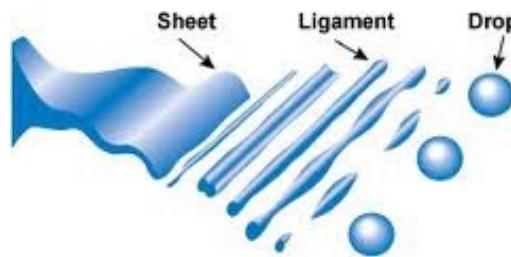
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Motivation – Ejecta processes

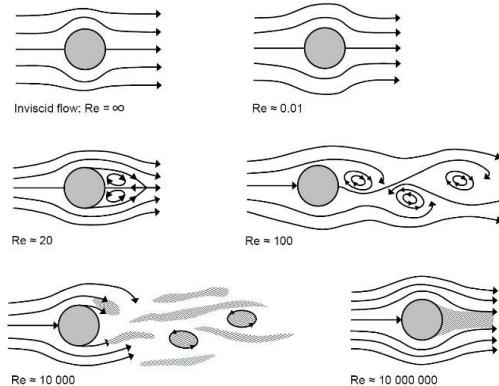
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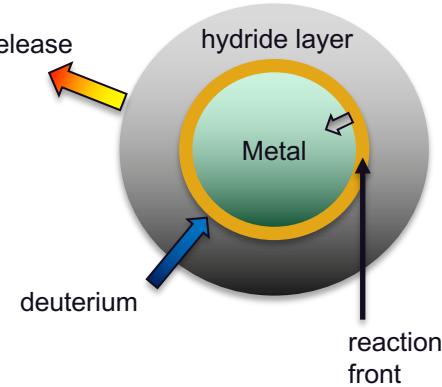
breakup



transport

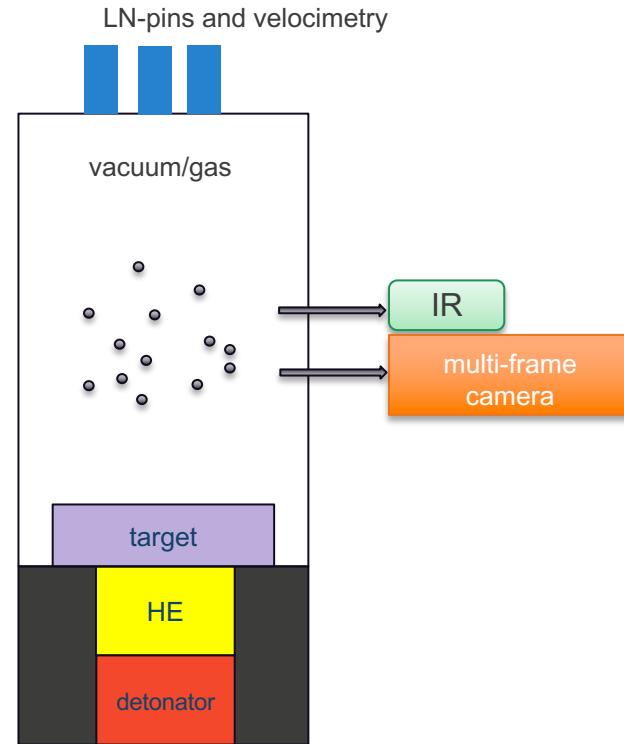
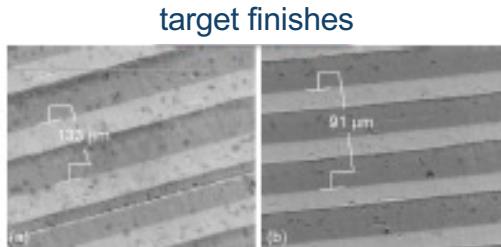


conversion



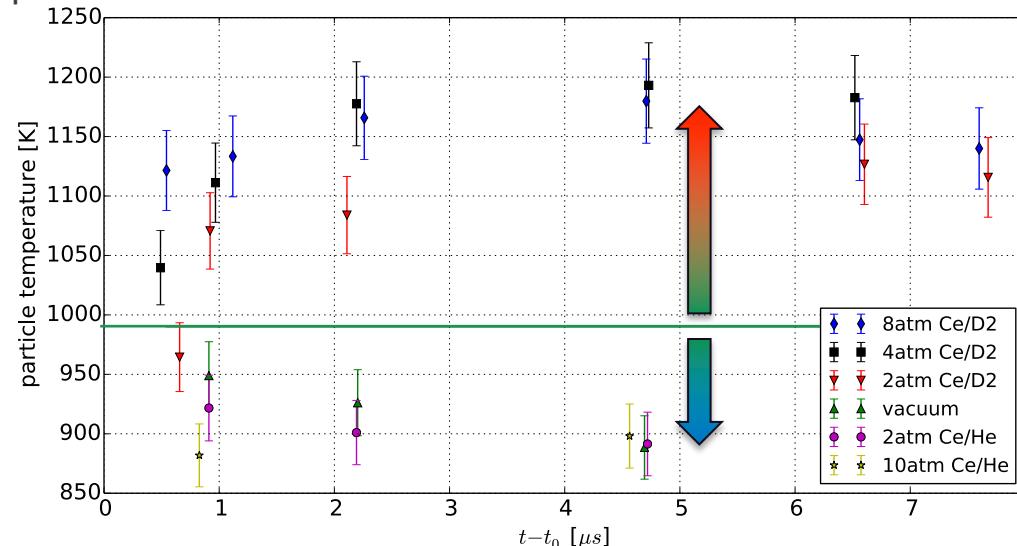
LDRD Experiment (PI: W. Buttler, LA-UR-19-21158)

- HE drives shock wave into target → cerium and tin ejecta into
 - Vacuum, He, D₂ at 2, 4, 8 atm
- IR imaging to measure radiance temperature
- PDV for ejecta velocities
- LN-pins for mass
- Initial ejecta temperature is 990K
- Mie scattering measures particle size
 - 12 micron mean diameter for cerium
 - 2 micron mean diameter for tin
- Inferred solid cerium ejecta particles



Radiance temperature observations from experiments

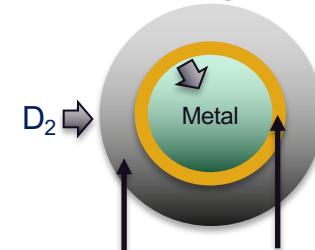
- Vacuum and helium media
 - Particle temperature drops (non-reactive)
- Deuterium medium
 - Particle temperature rises 200 K above initial temperature for $t < 5$ microseconds
 - Exothermic reaction
 - Maximum temperature reached after ~ 5 microseconds
 - Temperature drops for $t > 5$ microseconds



Physical assumptions for model development

- Assume liquid ejecta droplets are hydrodynamically stable or solid
- Exothermic chemical reactions occur at particle surface
 - Reaction is diffusion-controlled → diffusion through hydride layer
 - Shell remains intact
- Heat transfer is dominated by convection
 - Radiation is small in high density gas with moderate temperature differences
 - Temperature inside particle is uniform → lumped capacitance model
 - Biot number = $0.15 \ll 1$

Growing Shell

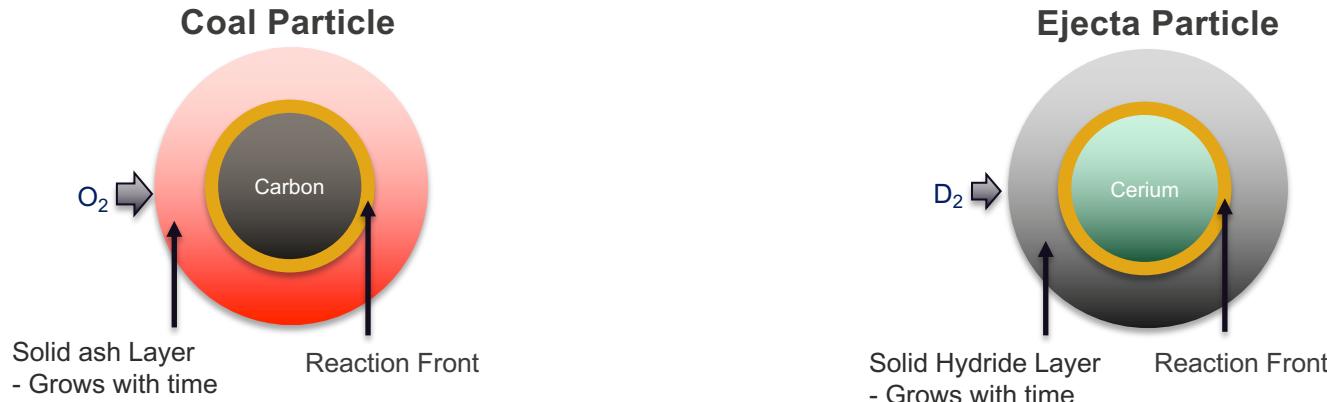


Solid Hydride Layer
- Grows with time

- Buttler LDRD report LA-UR-19-21158
- Buttler et al., Ejecta Transport, Breakup and Conversion, J. Dynamic Behavior Materials, 2017.

Coal combustion analog for solid core

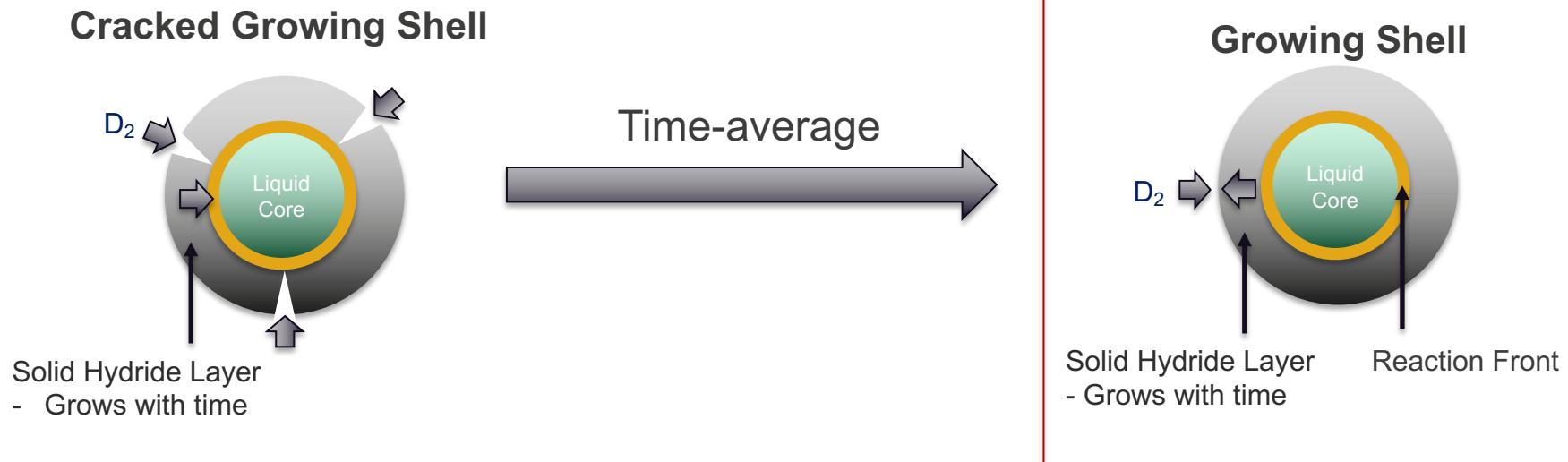
- High ash content coal combustion models are very similar*



- Ash layer quickly forms on surface of particle
 - Oxygen diffusion through ash layer controls carbon oxidation rate
 - Thickness of ash layer grows with time
-
- I. Glassman, Combustion, Ed. 3, 1996

Aluminum oxidation analog for liquid core

- Aluminum particles oxide in a similar fashion
- Oxide crust cracks → exposes fresh aluminum
 - Rapid kinetics based reaction “heals” cracks*
- Simplify by using effective diffusion coefficient → largest uncertainty



Mass Conservation

- D₂ diffuses through CeD₂ layer
- Quasi-steady diffusion through particle (lumped capacitance)

$$\frac{d}{dr} \left(r^2 \frac{dY}{dr} \right) = 0 \quad \Rightarrow \quad Y(r) = Y_\infty \frac{ab}{a-b} \left(\frac{1}{r} - \frac{1}{a} \right)$$

- Deuterium diffusion rate through particle $\dot{m}_{D_2} = \mathcal{D} 4\pi ab \frac{\rho_\infty Y_\infty}{b-a}$

- Cerium consumption rate $\text{Ce} + \text{D}_2 \rightarrow \text{CeD}_2$



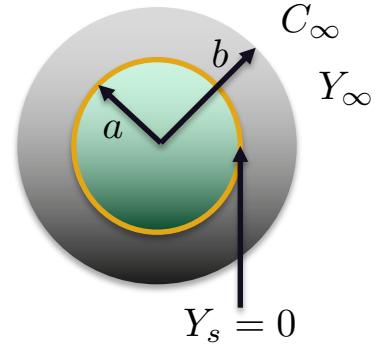
- Heat release rate

$$\dot{Q}_r = \dot{m}_{Ce} h_f$$

- Assume outer radius remains relatively constant

- Rate of change of inner cerium mass $\dot{m}_{Ce} = \rho_{Ce} 4\pi a^2 \frac{da}{dt}$

- Obtain ODE for inner radius $\frac{da}{dt} = i \frac{\rho_\infty}{\rho_{Ce}} \frac{Y_\infty \mathcal{D}}{b-a} \frac{b}{a}$



Energy Conservation

- Use lumped capacitance model
 - Thermal conduction inside particle faster than across gas/solid interface
 - Temperature is constant inside particle
- Convective heat transfer at surface

$$\dot{Q}_c = Nu\pi k_c 2b(T_\infty - T_p)$$

- Ranz-Marshall correlation (1952) for forced convection

$$Nu = 2 + 0.6Re_r^{1/2}Pr^{1/3}$$

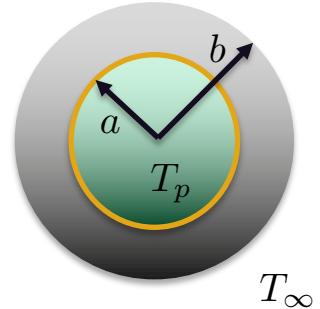
- Energy rate of change for particle

$$\dot{E}_p = \dot{Q}_r + \dot{Q}_c$$

↑ Reaction ↗ Convection

- Energy contained inside particle

$$\dot{E}_p = m_p c_p \frac{dT_p}{dt}$$



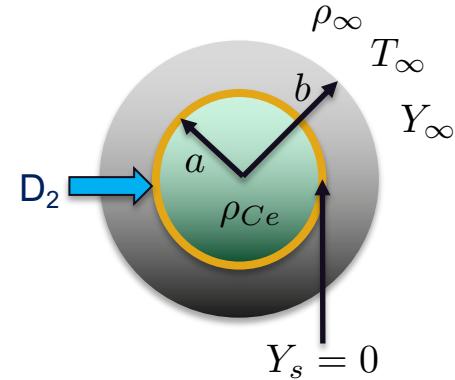
Model Summary

- Mass conservation

$$\frac{da}{dt} = i \frac{\rho_\infty}{\rho_{Ce}} \frac{Y_\infty \mathcal{D}}{b-a} \frac{b}{a}$$

$$i = \frac{W_{D_2}}{W_{Ce}}$$

$$\dot{m}_{Ce} = i 4 \pi a b \mathcal{D} \frac{\rho_\infty Y_\infty}{b-a}$$



- Energy conservation

$$\frac{de_p}{dt} = \frac{Nu}{2} \frac{c_p}{\tau_T} (T_\infty - T_p) + \frac{\dot{m}_{Ce} h_f}{m_p}$$

↑
Convective heat transfer ↑
Chemical heat release

$$\tau_T = \frac{\rho_p c_p (2b)^2}{12 k_c}$$

Thermal relaxation time

- Momentum conservation

- Focus on early time → constant velocity

Numerical Implementation

Numerical Implementation

- Initial reaction rate is fast
 - Potential division by zero

$$\frac{da}{dt} = i \frac{\rho_\infty}{\rho_{Ce}} \frac{Y_\infty \mathcal{D}}{b-a} \frac{b}{a}$$

- Use $n+1/2$ to prevent division by zero
- Naive discretization of reaction radius equation is inefficient
 - Accuracy highly dependent on timestep size
 - Numerical integration requires $O(10^5)$ timesteps per particle reaction time
 - Timestep can be much smaller than hydro timestep
- Seek analytical solution for mass conservation equation

Analytical Solution - Transformation

- Assume that all parameters are constant except the reaction radius a
- Integrate to obtain

$$1 - 3 \left(\frac{a}{b} \right)^2 + 2 \left(\frac{a}{b} \right)^3 = \frac{t}{\tau_r}$$

- Reaction time:

$$\tau_r = \frac{1}{6i} \frac{\rho_{Ce}}{\rho_\infty} \frac{b^2}{\mathcal{D}Y_\infty}$$

- Cubic equation:

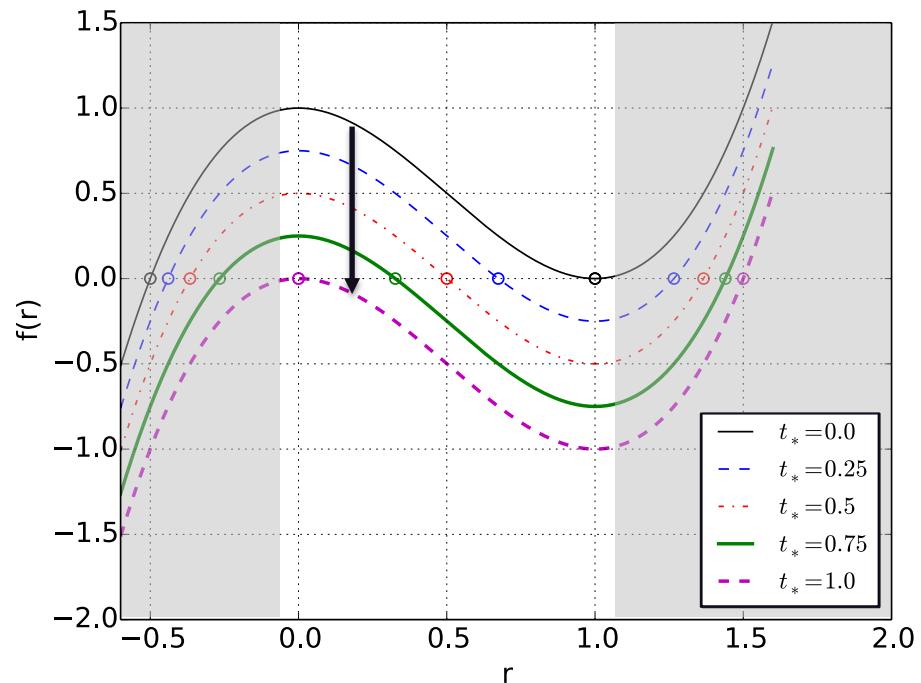
$$0 = 2r^3 - 3r^2 + (1 - t_*) \quad t_* = t/\tau_r \quad r = a/b$$

- Solution validity:

$$t_* \in [0, 1] \quad r \in [0, 1]$$

Graphical representation of solution

- Valid within $r \in [0, 1]$
- Solution starts at $r = 1$
- Moves left with increasing t_*
- Final value at $r = 0$
- Middle roots are valid



Analytical Solution

- Transform into depressed cubic: $0 = 2y^3 - \frac{3}{2}y + \frac{1-t_*}{2}$ $y = r + 1/2$

- Roots: $r_1 = \cos \theta + \frac{1}{2}$

$$r_2 = \cos\left(\frac{2\pi}{3} - \theta\right) + \frac{1}{2} \quad \theta = \frac{1}{3} \arccos(2t_* - 1)$$

$$r_3 = \cos\left(\frac{2\pi}{3} + \theta\right) + \frac{1}{2}$$

- Solution: $r(t_*) = \cos\left(\frac{2\pi}{3} - \theta(t_*)\right) + \frac{1}{2}$ $\theta(t_*) = \frac{1}{3} \arccos(2t_* - 1)$

Numerical Implementation

- At end of previous timestep $t_* = t_*^n \rightarrow t_*^{n+1} = t_*^n + \Delta t_*$
- Radius at t_*^n $r^n = r(t_*^n) = \cos\left(\frac{2\pi}{3} - \theta^n\right) + \frac{1}{2}$
- Radius at t_*^{n+1} $r^{n+1} = r(t_*^{n+1}) = \cos\left(\frac{2\pi}{3} - \theta^{n+1}\right) + \frac{1}{2}$
- Express new radius as radius change

$$\Delta r_n^{n+1} = r^{n+1} - r^n = \cos\left(\frac{2\pi}{3} - \theta^{n+1}\right) - \cos\left(\frac{2\pi}{3} - \theta^n\right)$$

Transform to original dimensional variables

- Dimensional reaction radius: $a^n = br^n$ $a^{n+1} = br^{n+1}$
- New reaction radius: $a^{n+1} = a^n + b\Delta r_n^{n+1}$
- Time nondimensionalization assumes τ_r const: $t_*^n = \frac{t^n}{\tau_r}$
- Accommodate variable properties by
 - Approximate as constant during timestep $\tau_r \rightarrow \tau_r^n$

$$\theta^n = \frac{1}{3} \arccos \left(2 \frac{t^n}{\tau_r^n} - 1 \right) \quad \theta^{n+1} = \frac{1}{3} \arccos \left(2 \left[\frac{t^n + \Delta t}{\tau_r^n} \right] - 1 \right)$$

Whole step discretization

$$a^{n+1} = a^n + b \Delta r_n^{n+1}$$



$$\Delta r_n^{n+1} = \cos\left(\frac{2\pi}{3} - \theta^{n+1}\right) - \cos\left(\frac{2\pi}{3} - \theta^n\right)$$



$$\frac{1}{3} \arccos\left(2\left[\frac{t^n + \Delta t}{\tau_r^n}\right] - 1\right) = \theta^{n+1}$$

$$\theta^n = \frac{1}{3} \arccos\left(2\frac{t^n}{\tau_r^n} - 1\right)$$

- Solution is exact for constant τ_r

Integration with energy equation

$$\frac{de_p}{dt} = \frac{Nu}{2} \frac{c_p}{\tau_T} (T_\infty - T_p) + \frac{\dot{m}_{Ce} h_f}{m_p}$$

- Energy equation is integrated with first order explicit scheme
 - Use average particle temperature and mass conversion rate at n+1/2

$$\frac{\Delta e_p}{\Delta t} = \frac{Nu}{2} \frac{c_p}{\tau_T} (T_\infty - T_p^{n+1/2}) + \frac{\dot{m}_{Ce}^{n+1/2} h_f}{m_p}$$

$$T^{n+1/2} = \frac{T_p^n + T_p^{n+1}}{2} \quad e^{n+1/2} = c_p T_p^{n+1/2}$$

$$\frac{\Delta e_p}{\Delta t} \left(1 + \frac{\Delta t}{\tau_T} \frac{Nu}{4} \right) = \frac{Nu}{2} \frac{c_p}{\tau_T} (T_\infty - T_p^n) + \frac{\dot{m}_{Ce}^{n+1/2} h_f}{m_p} \quad e^{n+1} = e^n + \Delta e$$

- Need mass conversion rate at n+1/2

Mass conversion rate at n+1/2

- Mass conversion rate: $\dot{m}_{Ce}^{n+1/2} = i4\pi a^{n+1/2} b \mathcal{D} \frac{\rho_\infty Y_\infty}{b - a^{n+1/2}}$
- Radius at n+1/2: $a^{n+1/2} = a^n + b \Delta r_n^{n+1/2}$
$$\Delta r_n^{n+1/2} = \cos\left(\frac{2\pi}{3} - \theta^{n+1/2}\right) - \cos\left(\frac{2\pi}{3} - \theta^n\right)$$
$$\theta^{n+1/2} = \frac{1}{3} \arccos\left(2 \left[\frac{t^n + \Delta t/2}{\tau_r^n} \right] - 1\right)$$

Obtain final state at n+1

- Find updated energy

$$\frac{\Delta e_p}{\Delta t} \left(1 + \frac{\Delta t}{\tau_T} \frac{Nu}{4} \right) = \frac{Nu}{2} \frac{c_p}{\tau_T} (T_\infty - T_p^n) + \frac{\dot{m}_{Ce}^{n+1/2} h_f}{m_p} \quad e^{n+1} = e^n + \Delta e$$

- Integrate radius another half step

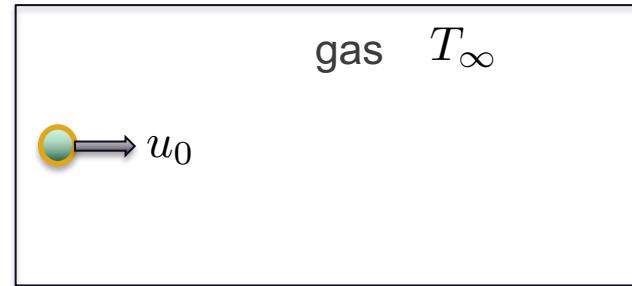
$$a^{n+1} = a^{n+1/2} + b \Delta r_{n+1/2}^{n+1}$$

$$\begin{aligned} \Delta r_{n+1/2}^{n+1} &= \cos\left(\frac{2\pi}{3} - \theta^{n+1}\right) - \cos\left(\frac{2\pi}{3} - \theta^{n+1/2}\right) \\ \frac{1}{3} \arccos\left(2 \left[\frac{t^{n+1/2} + \Delta t/2}{\tau_r^n} \right] - 1\right) &= \theta^{n+1} \\ \theta^{n+1/2} &= \frac{1}{3} \arccos\left(2 \left[\frac{t^n + \Delta t/2}{\tau_r^n} \right] - 1\right) \end{aligned}$$

Verification, Validation and Performance

Model verification

- Setup simple verification problem
 - Single particle inside domain
 - Constant velocity
 - Constant gas temperature



- Reaction radius ODE → constant diffusion coefficient

$$\frac{da}{dt} = i \frac{\rho_\infty}{\rho_{Ce}} \frac{Y_\infty \mathcal{D}}{b-a} \frac{b}{a}$$

- Energy equation

- Verified each term independently
- Adiabatic heat release
- Thermal relaxation

$$\frac{de_p}{dt} = \frac{Nu}{2} \frac{c_p}{\tau_T} (T_\infty - T_p) + \frac{\dot{m}_{Ce} h_f}{m_p}$$

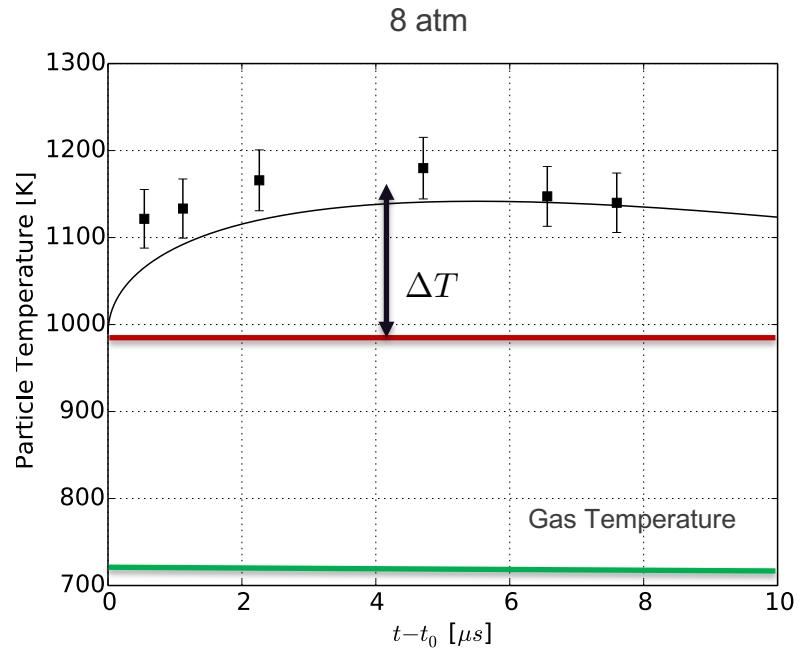
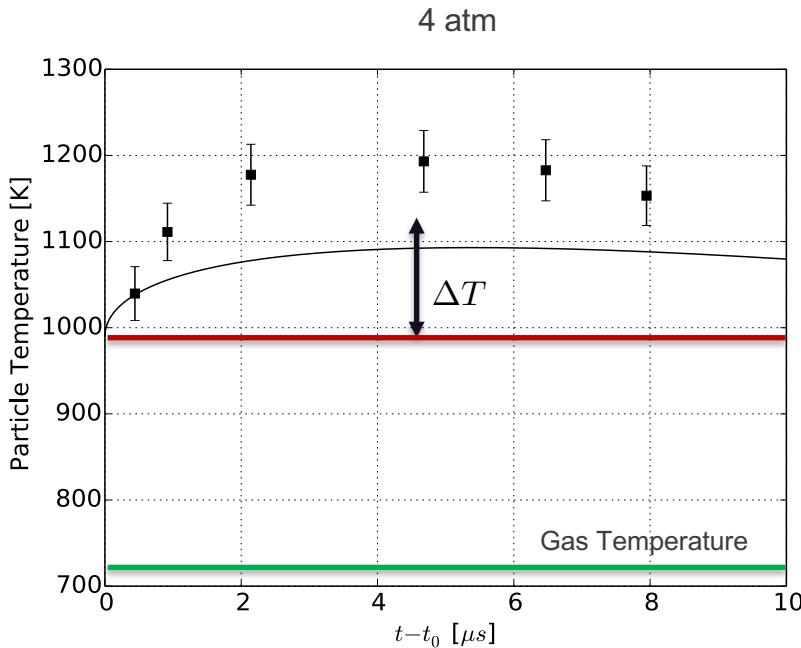
Validation – Change to post-shock conditions

- Tested model with Ce experiment at 4 and 8 atm pre-shock conditions

Initial Conditions	Initial Gas Pressure	4	8	atm
	Shocked Gas Temperature	720	740	K
	Shocked Metal Temperature	990	990	K
	Shocked Gas Density	2.3	4.5	kg/m ³
	Relative Velocity	480	470	m/s

Gas Properties		Metal Material Properties	
Specific heat	7.25 kJ/kg-K	Specific heat	269 J/kg-K
Thermal conductivity	0.138 W/m-K	Particle diameter	13 micron
Viscosity	1.72E-5 Pa-s	Particle density	6.7 g/cc
Specific heat ratio	1.4	CeD ₂ heat of formation	210 kJ/mol
		Diffusion coefficient	$\mathcal{D}(T_p) = 2.083589\text{E-}14 \cdot T_p^{1.941438}$

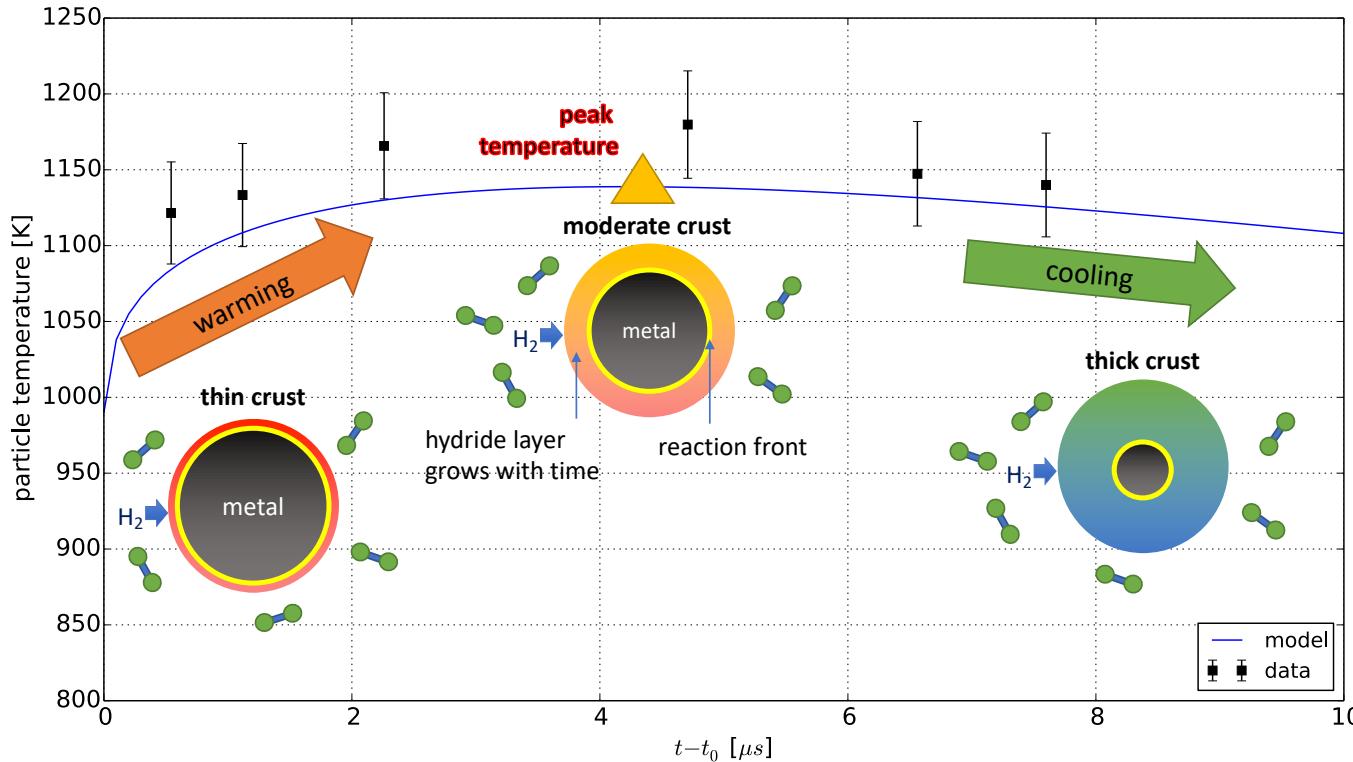
Particle temperature comparison



- Model roughly captures the 200 K increase in particle temperature
- Predicted temperatures are low for 4 atm case
- Relatively accurate for 8 atm case

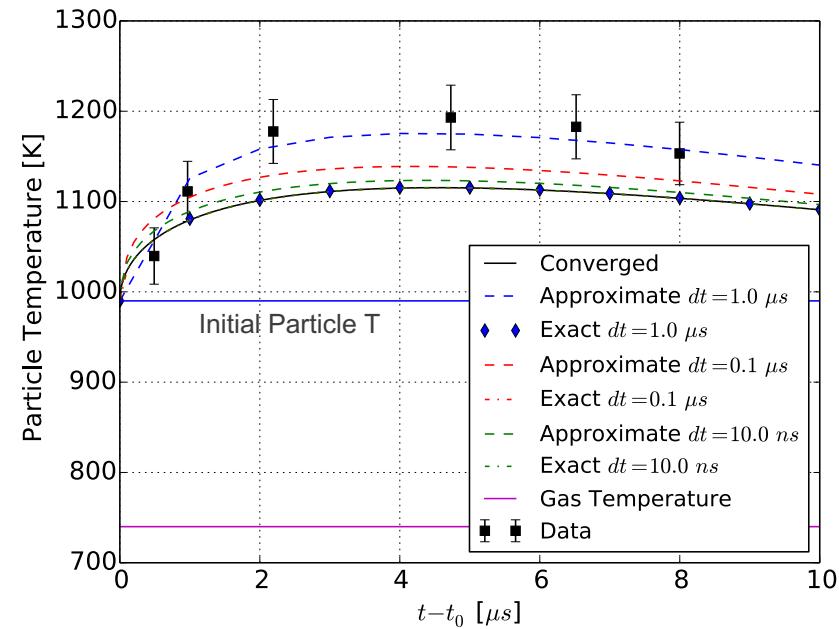
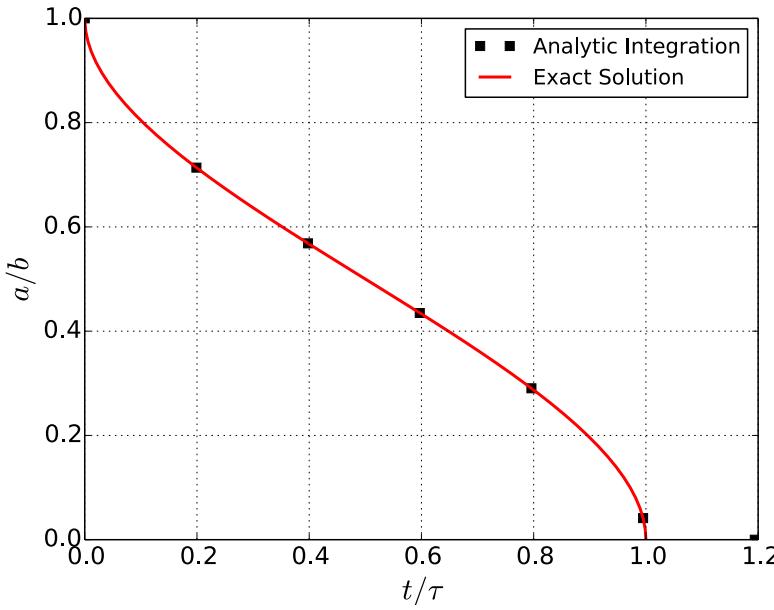
Competing processes

- 8 atm data
- Physical assumptions seem reasonable



Resolution sensitivity

- Reaction radius is exact regardless of resolution
- Temperature accurate regardless of resolution for exact scheme
 - First order approximate scheme overestimates temperature
- Both schemes converge to same solution

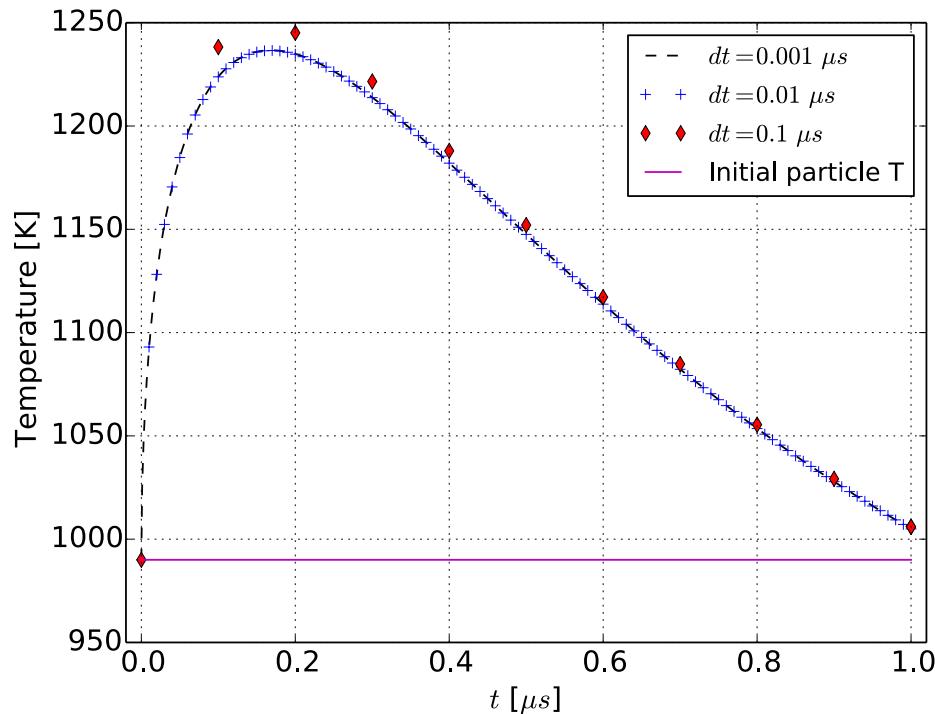
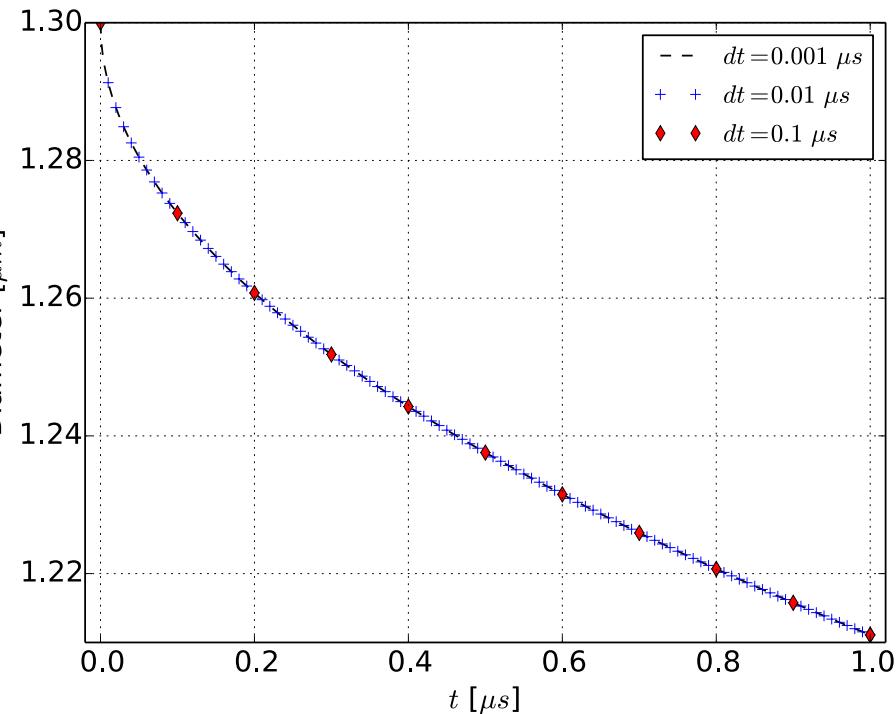


Model performance with small particles

- Particles in experiments are 13 microns in diameter → large
- Test model with 1.3 and 0.13 micron particles

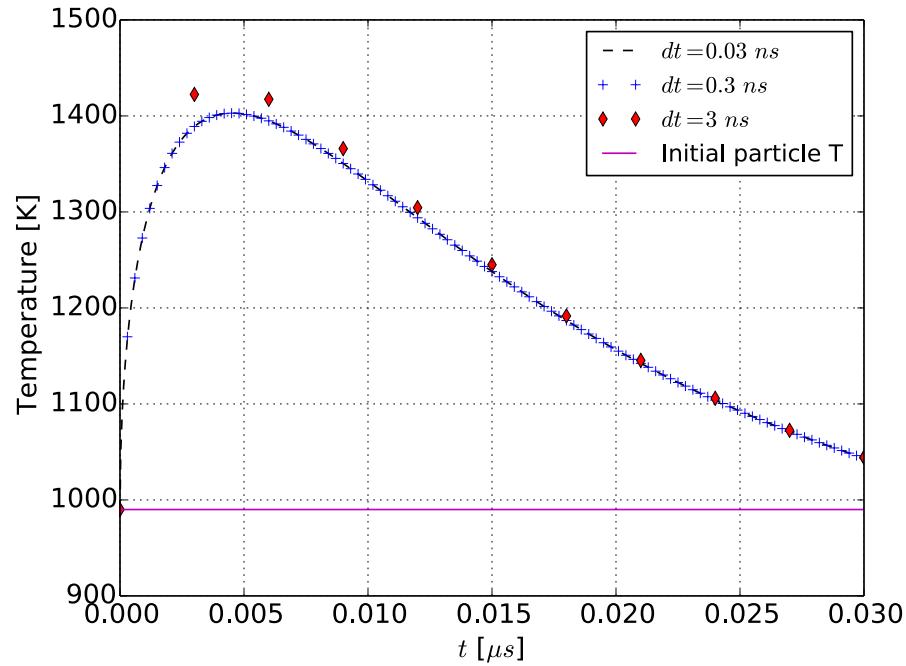
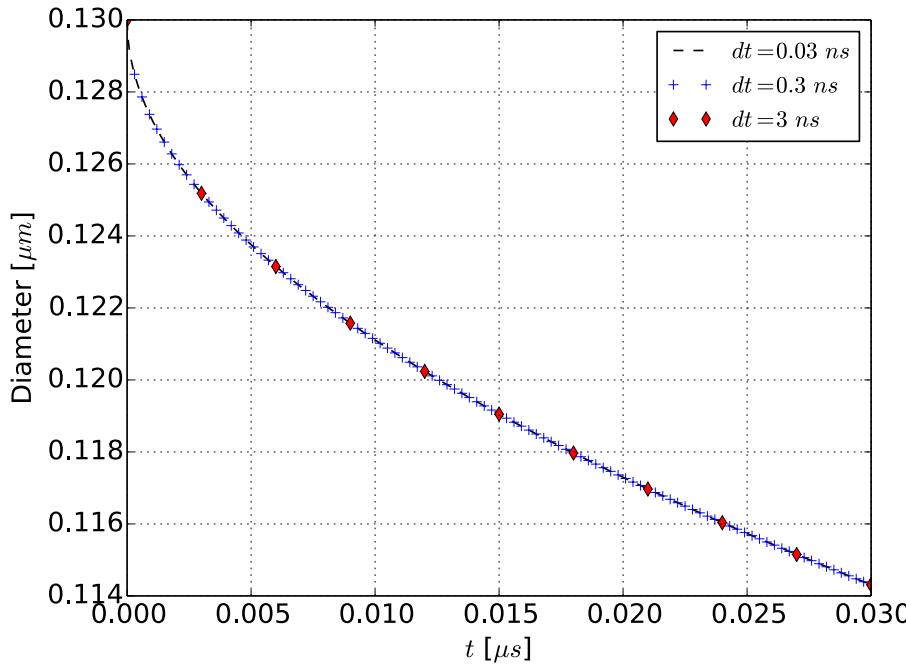
1.3 micron particles

- Reaction radius independent of timestep size
- Temperature slightly overestimated for largest timestep



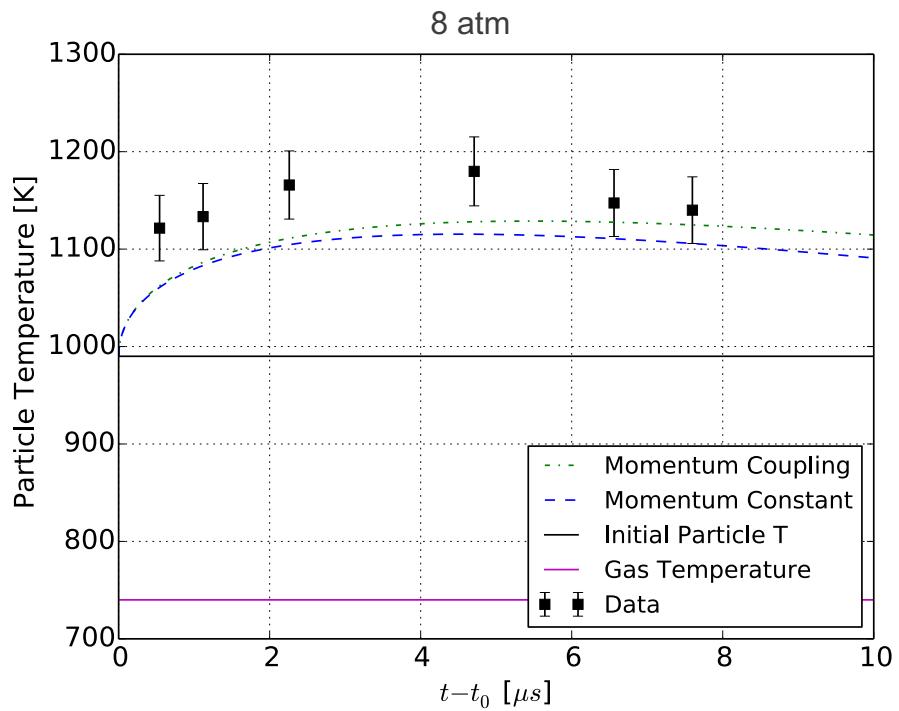
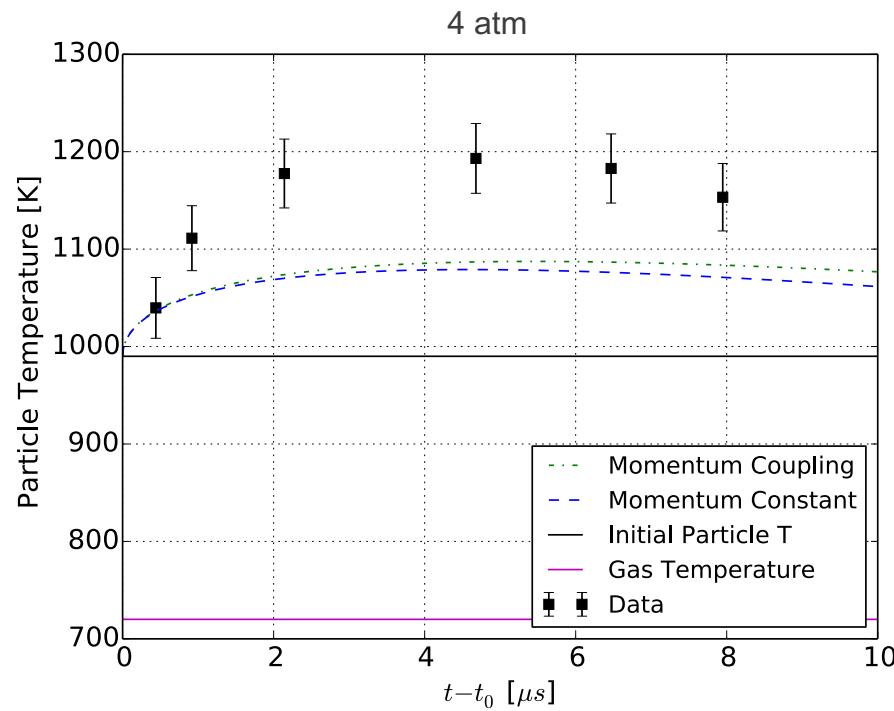
0.13 micron particles

- Reaction radius independent of timestep size
- Smaller timesteps at beginning increase accuracy
- May need adaptive timestep



Momentum Coupling effects

- Momentum coupling slows particles
 - Reduces heat conduction → increases particle temperature



Summary

- Simple diffusion-controlled reaction model
- Roughly approximates temperature increase of cerium ejecta particles
 - No tuning of parameters
 - Greatest uncertainty exist in diffusion coefficient
- Need more detail to accurately capture temperature rise
- Exact solution increases accuracy and minimizes computational cost
 - Both reaction radius and particle temperature
- Particle temperatures are reasonably accurate for range of sizes
 - Adaptive time-stepping may be required for greater accuracy

